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Cosmochemistry Program

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Introduction

The discovery of presolar grains in meteorites is one of the most exciting recent developments in meteoritics. Six types of presolar grain have been discovered: diamond, SiC, graphite, Si₃N₄, Al₂O₃ and MgAl₂O₄. These grains have been identified as presolar because their isotopic compositions are very different from those of Solar System materials. Comparison of their isotopic compositions with astronomical observations and theoretical models indicates most of the grains formed in the envelopes of highly evolved stars. They are, therefore, a new source of information with which to test astrophysical models of the evolution of these stars. In fact, because several elements can often be measured in the same grain, including elements that are not measurable spectroscopically in stars, the grain data provide some very stringent constraints for these models.

Our primary goal is to create large, unbiased, multi-isotope databases of single presolar SiC, Si_3N_4 , oxide and graphite grains in meteorites, as well as any new presolar grain types that are identified in the future. These will be used to: (i) test stellar and nucleosynthetic models, (ii) constrain the galactic chemical evolution (GCE) paths of the isotopes of Si, Ti, O and Mg, (iii) establish how many stellar sources contributed to the Solar System, (iv) constrain relative dust production rates of various stellar types and (v) assess how representative of galactic dust production the record in meteorites is. The primary tool for this project is a highly automated grain analysis system we have developed for the Carnegie 6f ion probe.

Progress

(1) Galactic chemical evolution of Si and Ti isotopes - Explanations for the range of Si isotopic compositions of mainstream SiC grains and O isotopic compositions of Group 1 and 3 oxide grains which appeal to galactic chemical evolution (GCE) have produced two paradoxical results: (1) most SiC grains appear to have come from stars which were initially more metal-rich (more chemically evolved) than the Sun, yet these stars must have formed hundreds of millions to billions of years before the Sun, and (2) most oxide grains found to date appear to have come from stars which were more metal-poor than the SiC parents despite the fact that the SiC source stars will also have produced considerable O-rich dust.

The validity of observations (1) and (2), as well as any explanations for them, rests on the accuracy of the galactic chemical evolution (GCE) model of (Timmes et al., 1995) (henceforth TWW95) used to estimate the metallicities. It has been known for some time that there is a discrepancy between the slope in a three isotope plot of the mainstream SiC Si isotope trend and the slope predicted by GCE models. We have used the C, Si and Ti isotopic compositions of 31 mainstream grains taken from the literature and data for 20 new 1-5µm Murchison grains measured by us to address several of the issues and explanations discussed above (Alexander and Nittler, 1998, 1999).

The non-solar normalization scheme proposed by (Clayton and Timmes, 1997) to explain the slope of the mainstream grains on a Si three isotope diagram predicts a Si GCE path that is hundreds of permil to the left of the mainstream grains. However, it requires the dredge-up of so much 12 C-rich He-shell material to explain the mainstream Si isotopic compositions that it can be ruled out on the basis of the grains' 12 C/ 13 C ratios. We also argue that the grains' C isotopic compositions require cool bottom processing on the RGB and that the grains probably came from stars of $\leq 2M_i$.

We have also used a X^2 fit to the Si and Ti isotope grain data to estimate the GCE paths of their minor isotopes as a function of metallicity. The best fit GCE paths in three isotope plots are generally sub-parallel to those of the solar normalized TWW95 model and, except for δ^{49} Ti and δ^{50} Ti, are quite well constrained. The biggest differences between the fitted evolution paths and those of the normalized TWW95 model involve δ^{30} Si and δ^{47} Ti. For several reasons, we argue that 30 Si is overproduced in the type II supernova models, explaining (i) its overproduction compared to 28,29 Si in the unnormalized TWW95 model, (ii) the shallower slope of the normalized TWW95 Si isotope GCE line compared to the mainstream grains, and (iii) the higher of δ^{29} Si/ δ^{30} Si ratio of the supernova Grains X and Si₃N₄ compared to the supernova models. 47 Ti appears to be underproduced in the normalized TWW95 model, presumably because it is underproduced in type II and/or type Ia supernova models.

Another potentially important result of the X^2 fit is that the estimated Si isotopic composition of the mean ISM at solar metallicity, [Fe/H]=0, is isotopically heavier than solar, δ^{29} Si≈80‰ and δ^{30} Si≈50‰. This suggests that the solar Si isotopic composition is somewhat atypical. The [Fe/H]=0 composition is slightly heavier than the mean of the SiC Si isotope distribution and if the SiC grains are compared to it rather than solar, the paradox of most mainstream grains being more metal-rich than the local ISM composition is largely resolved. Using the fit result as the [Fe/H]=0 reference composition results in a SiC parent star metallicity distribution which is closer to that inferred from the presolar oxide grains. A slight depletion of the Solar System Si isotopic composition compared to the local ISM can be achieved by addition of a few percent of material from a type II supernova, although the range of progenitor masses is quite limited if dramatic changes in the compositions of other elements like Ti and O are to be prevented.

(2) Stellar diffusion re-examined - Despite the explanation offered by the fit for the apparently super-solar metallicity of most SiC parent stars, we felt it important to examine the diffusing star scenario proposed by (Clayton, 1997). Radial orbital diffusion of stars in the Galaxy has been proposed as an explanation for the wide range of metallicities exhibited by stars of similar age at any given galactocentric distance. Clayton (1997) extended this idea by suggesting that, because the densities of stars and the molecular clouds which scatter them increase towards the galactic center, the overall radial diffusion would be outwards. Since the

inner regions of the Galaxy are also more metal-rich, the outwardly scattered stars that produced presolar grains would be more metal-rich than the local ISM material from which the Sun formed.

We have modeled this diffusion of stars, but also including the effect of the higher number density of stars and molecular clouds in the inner Galaxy. Using astronomical estimates for the distribution of AGB stars and molecular clouds as a function of galactocentric radius, we obtain a remarkably good fit to the stellar metallicity versus mass relationship inferred from the O isotopic compositions of the presolar oxide grains (Nittler and Alexander, 1998a, 1999a). However, the model does not predict that a majority of grains should come from AGB stars with metallicities higher than solar, unless an implausibly steep distribution of molecular clouds with galactic radius is assumed (Nittler and Alexander, 1999a). Thus, if the Si isotopes of presolar SiC grains indeed indicate that they formed in stars of higher than solar metallicity, it seems unlikely that radial diffusion of stellar orbits is a viable explanation.

(3) Successful development of the automated search and analysis technique

We has completed the main phase of development of the technique and the first results for ~250 presolar SiC grains analyzed automatically in a 24hr run (Nittler and Alexander, 1998b). We have also conducted a search for oxide grains in a mixed residue prepared from residues of Semarkona, Bishumpur and Krymka (Nittler and Alexander, 1999b). A total of 3000 grains were analyzed, and of these 24 proved to be presolar. As expected, many more presolar grains with near solar compositions were found compared to previous studies using the low-resolution direct imaging technique. However, the results revealed a much stronger than expected bias in the direct imaging method for detection of extreme compositions, such as Group 2 grains. This result clearly demonstrates the importance of our new method for acquiring unbiased isotopic data. In the course of this work, we also found what is likely the first presolar TiO₂ grain, adding yet another to the list of known presolar minerals. We have also found the first supernova oxide grain (Nittler et al., 1998).

(4) Development of large-radius multicollector SIMS

Funds from this grant, along with a supplemental PIUP grant (NAG5-10656), were used for the development of a large-radius mass spectrometer for our Cameca 6f ion microprobe. Construction of the CIW-built large-radius mass spectrometer began in early 2000 with installation of the 100 cm-radius magnet and its power supply in the ion microprobe lab. Assembly and testing of the magnetic sector vacuum housing was completed in late 2000. Using a thermal ionization source borrowed from a decommissioned TIMS instrument, the ion optics testing and modification of the magnet sector proceeded until the fall of 2001. Fabrication of the 75 cm-radius spherical electrostatic analyzer (ESA) was completed in early 2002 and tested with the thermal ionization source. Testing of the full double-focussing mass spectrometer (ESA+magnet) was done using a variable-energy modification to the thermal ionization source,

and the full mass spectrometer was demonstrated to achieve both mass and energy focussing over the full length of its 50 cm focal plane.

In June 2002 the CIW-built large radius mass spectrometer was permanently attached to the existing Cameca 6f ion probe, and since this time all ion optics testing has been achieved using the sputtered ion beam originating from the transfer optics of the 6f. During this time, the mass spectrometer has demonstrated an optical magnification of unity (source slit to collector slit) and a mass resolving power of 5400 (90%-10% definition, see Fig. 2) without reducing the sputtered beam size with slits. The magnet power supply maintains a short-term current stability of 15 ppb, equivalent in mass to 1 part in 20,000 at oxygen; this short-term stability is maintained over the long-term by a magnet regulation algorithm using a NMR-based measurement of the magnetic field. Software for computer control of the NMR and magnet are completed.

Fabrication of the first-generation multicollector is 75% completed. It consists of five channels, four of which are moveable under vacuum and can accept either an ETP 17-stage electron multiplier (EM) or a shielded ceramic Faraday cup. The central fixed-position channel will have a deflector-selectable routing of the beam to one of three detectors (EM, FC or channel plate). Pulse counting measurements with the EMs will be made with Phillips amplifiers and discriminators coupled to Agilent counters. All detectors, measurement electronics and software are in-hand and waiting for completion of the multicollector track assembly.

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